

Annealing-tuned Hall coefficient in single crystals of the YbNi₂B₂C heavy fermion

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Abstract

We present temperature-dependent magneto-transport measurements on as-grown and annealed YbNi₂B₂C single crystals. Annealing causes drastic changes in the Hall coefficient, $R_H(T)$. Whereas for as-grown samples the Hall coefficient is negative between room temperature and 2 K, with a pronounced *minimum* at ≈ 22 K, for the samples annealed at 950° C for 150 hours, $R_H(T)$ changes its sign twice in the same temperature range: from negative to positive on cooling below ~ 100 K and back to negative below ~ 10 K, and has a clear *maximum* at ≈ 45 K. Intermediate temperature dependencies can be achieved by reducing the annealing time. These findings are discussed within the framework of an annealing dependence of the skew scattering in conjunction with the recent structural, thermodynamic and transport studies of the effects of annealing in YbNi₂B₂C.

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I. INTRODUCTION

$\text{YbNi}_2\text{B}_2\text{C}$ is the heavy fermion [1, 2] member of a rich and complex family of quaternary borocarbibides, $\text{RNi}_2\text{B}_2\text{C}$ (R = rare earth). This family encompasses a wide range of physical phenomena: interaction between local moment magnetism and superconductivity, peculiar, highly anisotropic, metamagnetism, and strong electronic correlations [3, 4]. Unique features of $\text{YbNi}_2\text{B}_2\text{C}$ such as moderately high electronic specific heat coefficient, $\gamma \approx 500 \text{ mJ/mol K}^2$, well separated characteristic temperatures/energy scales (no superconductivity or long-range magnetic order above 0.03 K, Kondo temperature, $T_K \approx 10 \text{ K}$ and crystal electric field splitting, $T_{CEF} \approx 100 \text{ K}$ [1, 5, 6, 7]), as well as its availability in a single-crystalline form, make it a model system for detailed studies of strongly correlated Yb-based materials. Recently it was shown [8] that whereas the effect of annealing on the resistivity of single crystals of non-magnetic ($\text{R} = \text{Y, Lu}$) and "good local moment" ($\text{R} = \text{Gd-Tm}$) members of the $\text{RNi}_2\text{B}_2\text{C}$ series is an ordinary decrease of the residual resistivity consistent with Matthiessen's rule, for $\text{YbNi}_2\text{B}_2\text{C}$ extraordinary annealing-induced changes were found in the whole studied temperature range, 2 - 300 K. A subsequent detailed investigation [9] delineated the effects of annealing on $\text{YbNi}_2\text{B}_2\text{C}$: thermodynamic properties (DC magnetic susceptibility, $\chi(T)$, and heat capacity, $C_p(T)$) remained practically unchanged, whereas the zero-field transport properties (resistivity, $\rho(T)$, and thermoelectric power, $S(T)$) showed dramatic changes. The evolution of the physical properties of $\text{YbNi}_2\text{B}_2\text{C}$ upon annealing was rationalized in terms of redistribution of local Kondo temperatures associated with ligand disorder for a small (on the order of 1%) fraction of the ytterbium sites. The nature of the ligand disorder was addressed by performing single crystal X-ray diffraction and transmission electron microscopy measurements [10]: lattice dislocations were found to be the dominant defect type in as-grown single crystals. These dislocations were suggested to be responsible for the environment changes around adjacent Yb^{3+} ions leading to a distribution of the local Kondo temperatures and were shown to be (at least partially) annealed out by heat treatment of the sample in vacuum at 950°C. Additionally, extended-temperature-range resistivity measurements (up to $\sim 1000 \text{ K}$) [10] suggested that this distribution of the local Kondo temperatures does not extend above $\sim 500 - 600 \text{ K}$.

The aforementioned publications [8, 9, 10] have clearly shown that the zero-field transport properties of $\text{YbNi}_2\text{B}_2\text{C}$ are exceptionally sensitive to very small (effecting on the order of

1% of the Yb^{3+} sites) perturbations of the structure. Structural defects such as these are not unique to $\text{YbNi}_2\text{B}_2\text{C}$, and their effects should be taken into account if any detailed theoretical modelling of the transport properties of materials with hybridizing moments is to be carried out.

Once $\text{YbNi}_2\text{B}_2\text{C}$ is established as a model system for the investigation of the effects of minor structural disorder on the physical properties of heavy fermion materials, it is of clear interest to extend the range of the properties examined. In this work we concentrate on a very common magneto-transport characteristic of materials: the Hall effect. It was realized quite early [11] that the temperature- and field- dependence of the Hall coefficient depends on the nature and purity of a material and can be quite complex. The temperature-dependent Hall coefficients in different heavy fermion compounds share common features (see *e.g.* [12, 13, 14] for a review): (i) $R_H(T)$ is very large, usually it is positive for Ce- and U- based systems and negative for Yb- based materials; (ii) with decreasing temperature $|R_H(T)|$ increases, passing through a maximum at a temperature of the order of a coherence temperature, $T_{max} \sim T_{coh}$, then rapidly decreasing and finally, at low temperatures (in the coherent state) approaching a constant value (or has additional features in the case of a superconducting or magnetically ordered ground state). Since the general behavior of the temperature-dependent Hall coefficient appears to be robust, with T_{coh} as the only relevant energy scale that is reflected as a feature in $R_H(T)$, it will be of interest to verify if in $\text{YbNi}_2\text{B}_2\text{C}$ annealing can modify $R_H(T)$ in a comprehensible manner.

In this work we present temperature-dependent Hall coefficient measurements on as-grown and annealed (to differing degrees) $\text{YbNi}_2\text{B}_2\text{C}$ single crystals and compare the results with effects of annealing on the non-magnetic, $\text{LuNi}_2\text{B}_2\text{C}$, analogue.

II. EXPERIMENTAL

Single crystals of $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ were grown by the high-temperature flux method using Ni_2B as a flux [1, 3, 15]. Several clean, well-formed and thin $\text{YbNi}_2\text{B}_2\text{C}$ crystals were chosen from the same batch. Small residual droplets of flux, when present, were polished off of the surfaces of the crystals. The crystals were cut with a wire saw into flat bars with lengths of 3-4 mm, widths of 1.5-2.5 mm and thicknesses of 0.15-0.2 mm. The lengths of each bar were approximately parallel to the [110] crystallographic direction. Five contacts were

attached to the samples using Pt wire with the Epotek H20E silver epoxy for simultaneous measurements of resistivity and Hall effect (see inset to Fig. 1). The temperature- and field- dependent resistivity, $\rho(H, T)$ and the Hall resistivity, $\rho_H(H, T)$, were measured using a four probe, ac technique ($f = 16$ Hz, $I = 1\text{-}3$ mA) in a Quantum Design Inc., Physical Property Measurement System (PPMS) instrument with a separate channel assigned to each measurement. The applied magnetic field was kept perpendicular to the electrical current for both, $\rho(H, T)$ and $\rho_H(H, T)$. To eliminate the effect of inevitable (small) misalignment of the voltage contacts, the Hall measurements were taken for two opposite directions of the applied field, H and $-H$, and the odd component, $(\rho_H(H) - \rho_H(-H))/2$ was taken as the Hall resistivity. After the measurements on the as-grown samples were finished the contacts were taken off from the surface of the samples, the samples were lightly polished to remove the residues of the silver epoxy and thoroughly cleaned with toluene and methanol, placed in a Ta foil envelope and then placed into a quartz insert of a high vacuum annealing furnace. Similarly to the procedure described in [8, 9, 10], the insert was continuously maintained at a pressure of less than 10^{-6} Torr during the annealing cycle that started with an ~ 1 hour dwell at 200°C followed by the desired time anneal at 950°C and then a cool-down (for 4-5 hours) to room temperature. New set of contacts was attached after annealing. Each sample was subjected to only one annealing cycle with the measurements for as-grown material performed for each specimen used for annealing. The same routine was used for the $\text{LuNi}_2\text{B}_2\text{C}$ crystals for which only one annealing time, 150 hours, was utilized.

III. RESULTS AND DISCUSSION

The zero field resistivity data from as-grown $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ and annealed at 950°C for 150 hours $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ samples are shown in Fig. 1(a), data for different annealing times at 950°C are presented in Fig. 1(b). The results for $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ including the evolution of the $\rho(T)$ curves for the intermediate annealing times for $\text{YbNi}_2\text{B}_2\text{C}$ are consistent with those reported in [8, 9, 10] showing the high reproducibility of the samples and the annealing procedure. All the measured as-grown $\text{YbNi}_2\text{B}_2\text{C}$ samples (Fig. 1(b)) have very similar resistivities, further suggesting very good reproducibility within the same batch and reasonable accuracy of the measurements of the samples' dimensions and contact positions. At first glance the data for $\text{LuNi}_2\text{B}_2\text{C}$ (Fig. 1(a)) seem not to obey the

Matthiessen's rule. We believe that in this case the apparent difference in the $\rho(T)$ slopes between as-grown and annealed sample is caused by somewhat higher than for the other samples (but still reasonable within the average size) accumulated error in the dimensions and contact position measurements (a 20-25% total correction would be required to have the $\rho(T)$ curves for the as-grown and annealed $\text{LuNi}_2\text{B}_2\text{C}$ data to be parallel to each other). If the room temperature slopes of the two $\rho(T)$ plots for $\text{LuNi}_2\text{B}_2\text{C}$ are normalized then the change associated with annealing is the simple Matthiessen's rule shift reported in previous work [8].

Fig. 2 shows selected isothermal, field-dependent Hall resistivity curves for several samples. Although for $\rho_H(H)$ taken at lower temperatures some curvature is observed, it is not large enough to significantly change the value or the behavior of the Hall coefficient as measured in different fields. A comparison between panels (a) and (b) clearly shows that for some temperatures the sign of the Hall resistivity is opposite for the as-grown and the annealed $\text{YbNi}_2\text{B}_2\text{C}$ samples, whereas at the base temperature and near the room temperature for both samples the Hall resistivity is negative.

The temperature-dependent Hall coefficients ($R_H(T) = \rho_H(T)/H$) of as-grown $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ and annealed at 950°C for 150 hours $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ are shown in Fig. 3. For $\text{LuNi}_2\text{B}_2\text{C}$ $R_H(T)$ is weakly temperature-dependent and practically unaffected by annealing; over the whole temperature range it is negative and has values between $-(2-4) \times 10^{-12} \Omega \text{ cm/Oe}$. These values and general behavior are consistent with the weakly temperature-dependent Hall coefficient $-(1-6) \times 10^{-12} \Omega \text{ cm/Oe}$ measured on polycrystalline samples of $\text{LuNi}_2\text{B}_2\text{C}$ and several other borocarbides ($\text{RNi}_2\text{B}_2\text{C}$, $\text{R} = \text{Y, La, Gd, Ho}$) [16, 17, 18, 19, 20]. The temperature-dependent Hall coefficient of as-grown $\text{YbNi}_2\text{B}_2\text{C}$ is negative over the whole 2-300 K temperature range, at room temperature its value is $R_H^{300K} \approx -5 \times 10^{-12} \Omega \text{ cm/Oe}$, this value decreases on cooling down, has a well-defined minimum at $\approx 22 \text{ K}$, with R_H^{min} slightly lower than $-30 \times 10^{-12} \Omega \text{ cm/Oe}$ and then increases reaching $\approx -11 \times 10^{-12} \Omega \text{ cm/Oe}$ at 2 K. This behavior is consistent with a model $R_H(T)$ behavior expected for an Yb-based heavy fermion. Curiously, the $\text{YbNi}_2\text{B}_2\text{C}$ sample annealed at 950°C for 150 hours appears to show very dissimilar behavior: its Hall coefficient is negative at room temperature ($R_H^{300K} \approx -1.5 \times 10^{-12} \Omega \text{ cm/Oe}$), it decreases slightly on cooling down, passes through very shallow minimum at about 175 K, increases on further cooling, crosses zero at $\sim 100 \text{ K}$, goes through maximum at $\approx 45 \text{ K}$ ($R_H^{max} \approx 6.5 \times 10^{-12} \Omega \text{ cm/Oe}$), and then decreases again.

cm/Oe), crosses zero again at ~ 10 K and reaches $\approx -7 \times 10^{-12} \Omega \text{ cm/Oe}$ at 2 K. So, as a result of annealing, a minimum in $R_H(T)$ appears to be transformed into a maximum with its position shifted ~ 20 K higher. The literature $R_H(T)$ data on a polycrystalline $\text{YbNi}_2\text{B}_2\text{C}$ sample shown negative Hall coefficient with no minima or maxima between 4.2 and 300 K [21].

The general picture of the temperature dependence of the Hall coefficient in heavy fermion materials has been presented in [22, 23, 24, 25] (see also [12, 13, 14] for more comprehensive reviews). Within this picture the temperature dependence of the Hall coefficient is the result of two contributions: a residual Hall coefficient, $R_H^{res} = \rho_H^{res}/H$, and a Hall coefficient due to the skew scattering, $R_H^s = \rho_H^s/H$. The residual Hall coefficient is ascribed to a combination of the ordinary Hall effect and residual skew scattering by non-hybridizing defects and impurities and, to the first approximation, is considered to be temperature-independent, although, realistically, both the ordinary Hall effect and the residual skew scattering may have weak temperature dependence. The temperature-dependent skew scattering contribution (R_H^s) at high temperatures ($T \gg T_K$, where T_K is the Kondo temperature) increases as the temperature is lowered in a manner that is mainly due to the increasing magnetic susceptibility. At lower temperatures R_H^s passes through a crossover regime, then has a peak at a temperature on the order of the coherence temperature, T_{coh} , and finally, on further cooling rapidly decreases (in the coherent regime of skew scattering by fluctuations) to zero (*i.e.* R_H ultimately levels off to the $\sim R_H^{res}$ value at very low temperatures [23, 24, 25]). Detailed theoretical descriptions of the temperature dependence of the skew scattering contribution to the Hall coefficient [22, 24] in the incoherent regime ($T > T_{coh}$) offered a general expression, $R_H^s = \gamma \tilde{\chi}(T) \rho(T)$, where $\tilde{\chi}(T)$ is a reduced susceptibility, and $\rho(T)$ is the resistivity due to the resonant scattering. In practice, $\tilde{\chi}(T) = \chi(T)/C$, where $\chi(T)$ is a temperature dependent susceptibility and C is the Curie constant and $\rho(T) = \rho_{experimental} - \rho_{phonon} - \rho_{residual}$. In this work the resonant (magnetic) scattering contribution to the resistivity can be approximated as $\rho_{magn} = \rho_{Yb}(T) - \rho_{Lu}(T)$, where $\rho_{Yb}(T)$ and $\rho_{Lu}(T)$ are the experimental temperature-dependent resistivities of the $\text{YbNi}_2\text{B}_2\text{C}$ and $\text{LuNi}_2\text{B}_2\text{C}$ respectively. The coefficient γ is different for two different temperature regimes: $T \gg T_K$ and $T_{coh} \leq T \leq T_K$ and depends on the details of scattering in different channels, a crossover region exists between these two temperature regimes. This was used to explain temperature dependence of several heavy fermion compounds (see *e.g.* [12, 13, 14]).

The temperature-dependent Hall coefficient and the product of magnetic susceptibility and the resonant scattering contribution to resistivity $[\chi(T)(\rho_{Yb}(T) - \rho_{Lu}(T))]$ for as grown and annealed at 950°C for 150 hours YbNi₂B₂C are shown in Fig. 4. For $\chi(T)$ the $H \parallel c$ data from [9] were used ($\chi(T)$ was shown [9] to be annealing-independent), for the estimate of $\rho_{magn}(T)$ the experimental curves were utilized, experimental $\rho_{Lu}(T)$ data for the annealed LuNi₂B₂C sample were utilized, the curve was extrapolated below the superconducting transition as $\rho = \rho_0 + AT^2$. There are striking similarities between respective $R_H(T)$ and $\chi(T)\rho_{magn}(T)$ curves. For the as-grown sample in both curves the position of the extremum (minimum in R_H and maximum in $\chi\rho_{magn}$) is approximately the same and in the 50-300 K range both temperature dependencies are rather steep. For the annealed sample the sign of the curvature of the $R_H(T)$ changes, the temperature dependence becomes slower, and a rather shallow maximum at T_{max} that is higher than T_{min} of the as-grown sample, is observed. It appears that (around and above T_{coh}) the product $\chi(T)\rho_{magn}(T)$ reproduces the main features observed in R_H provided the coefficient γ in both regimes has a negative sign. The similarity becomes even greater if we assume that there is a substantial, weakly temperature-dependent, possibly annealing-dependent, residual Hall effect contribution, R_H^{res} to the measured Hall coefficient. Then the apparent change of sign in the measured temperature-dependent Hall coefficient for the annealed sample can be simply a result of an "offset" caused by the R_H^{res} contribution with the skew scattering contribution being negative and approximately following $R_H^s = \gamma\tilde{\chi}(T)\rho_{magn}(T)$ (except the low temperature, coherent, region) theoretical description.

The same analysis can be extended to the intermediate annealing times at 950°C. $R_H(T)$ and $(-\chi(T)\rho_{magn}(T))$ for as grown and annealed at 950°C for 5, 20, 48, and 150 hours YbNi₂B₂C are shown in Fig. 5. Similar to the discussion above for the two extreme cases, all salient features of the $R_H(T)$ curves (Fig. 5(a)) are quite accurately reproduced in the respective $-\chi(T)\rho(T)_{magn}$ curves which, as indicated, are plotted with negative sign, for more transparent comparison.

The dramatic changes in $R_H(T)$ for as-grown and annealed YbNi₂B₂C can be fully understood within the framework of the existing models of the Hall effect in heavy fermions [22, 24] and our earlier annealing studies [8, 9, 10]. Since the magnetic susceptibility of YbNi₂B₂C does not change with annealing [9], R_H^s follows the changes in resistivity that were attributed [9, 10] to the annealing-induced redistribution of local Kondo temperatures

associated with a ligandal disorder for a small number of ytterbium sites. No additional, Hall effect specific, mechanism seems to be required to qualitatively understand the drastic changes in experimentally measured temperature-dependent Hall coefficient.

Whereas the temperature-dependent Hall coefficient for as-grown $\text{YbNi}_2\text{B}_2\text{C}$ perfectly fits the general prejudice for what should be expected for a Yb-based heavy-fermion material, we have shown that it is actually a consequence of the ligandal disorder for a small number of the Yb-sites. Similarly to the case of zero-field resistivity [10], it should be stressed that any detailed comparison of the theoretical models with experimental data for the Hall effect in heavy fermions should take into account this extreme sensitivity of the Hall coefficient (*via* the resonant scattering part of resistivity) to the small local disorder.

IV. SUMMARY

We have measured the temperature dependent Hall coefficient of samples of as-grown and annealed at 950°C for 5-150 hours $\text{YbNi}_2\text{B}_2\text{C}$ single crystals as well samples of as pure as-grown and annealed $\text{LuNi}_2\text{B}_2\text{C}$ crystals. Whereas annealing has very little effect on $R_H(T)$ of $\text{LuNi}_2\text{B}_2\text{C}$, the Hall coefficient of $\text{YbNi}_2\text{B}_2\text{C}$ changes drastically upon annealing. The changes in the measured $R_H(T)$ of $\text{YbNi}_2\text{B}_2\text{C}$ can be understood within a model for the skew scattering contribution to the Hall coefficient in the incoherent regime [22, 24] with some, possibly weakly temperature-dependent, residual contribution to the Hall coefficient. The changes in $R_H(T)$ of $\text{YbNi}_2\text{B}_2\text{C}$ upon annealing are directly connected with the changes in zero field resistivity that were attributed to the redistribution of local Kondo temperatures for a small number of ytterbium sites [9, 10].

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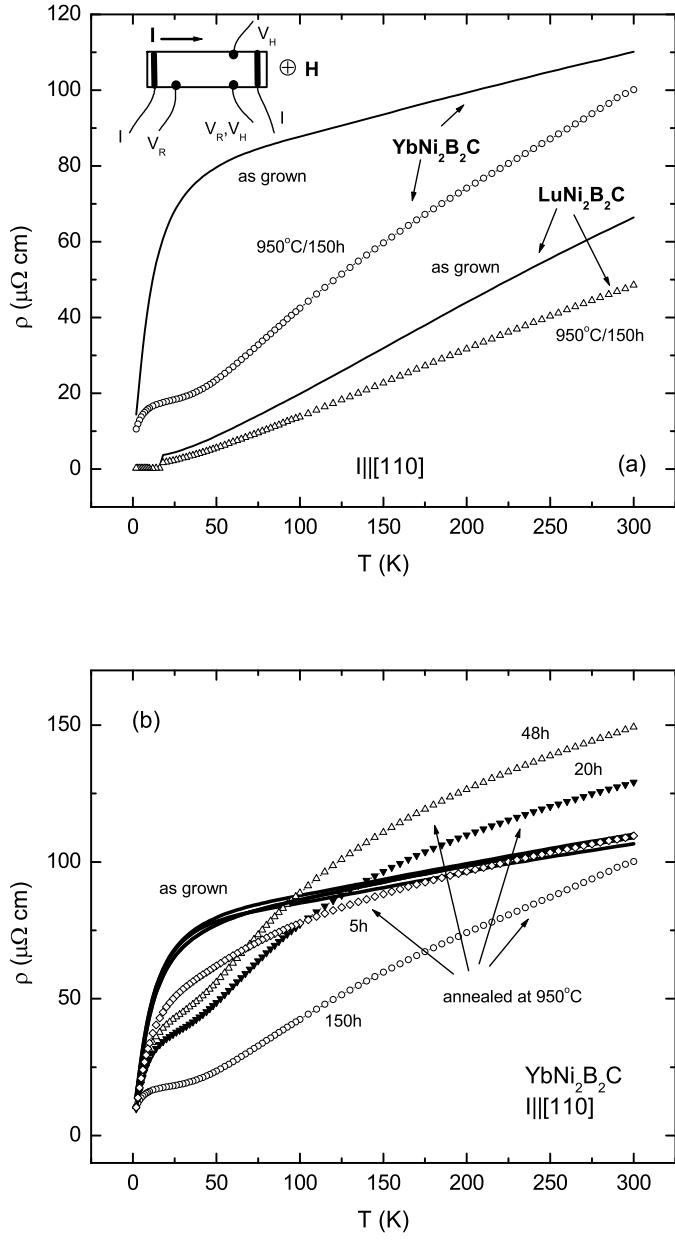


FIG. 1: (a) Zero-field resistivity of as-grown $\text{YbNi}_2\text{B}_2\text{C}$ (line) and $\text{LuNi}_2\text{B}_2\text{C}$ (line) and annealed at 950°C for 150 hours $\text{YbNi}_2\text{B}_2\text{C}$ (\circ) and $\text{LuNi}_2\text{B}_2\text{C}$ (\triangle) samples. Inset: sketch of the contact arrangement for simultaneous resistivity and Hall effect measurements (subscripts R and H respectively on the sketch). (b) Zero-field resistivity of four as-grown $\text{YbNi}_2\text{B}_2\text{C}$ crystals (lines), and the same samples annealed at 950°C for 150 (\circ), 48 (\triangle), 20 (\blacktriangledown) and 5 (\diamond) hours.

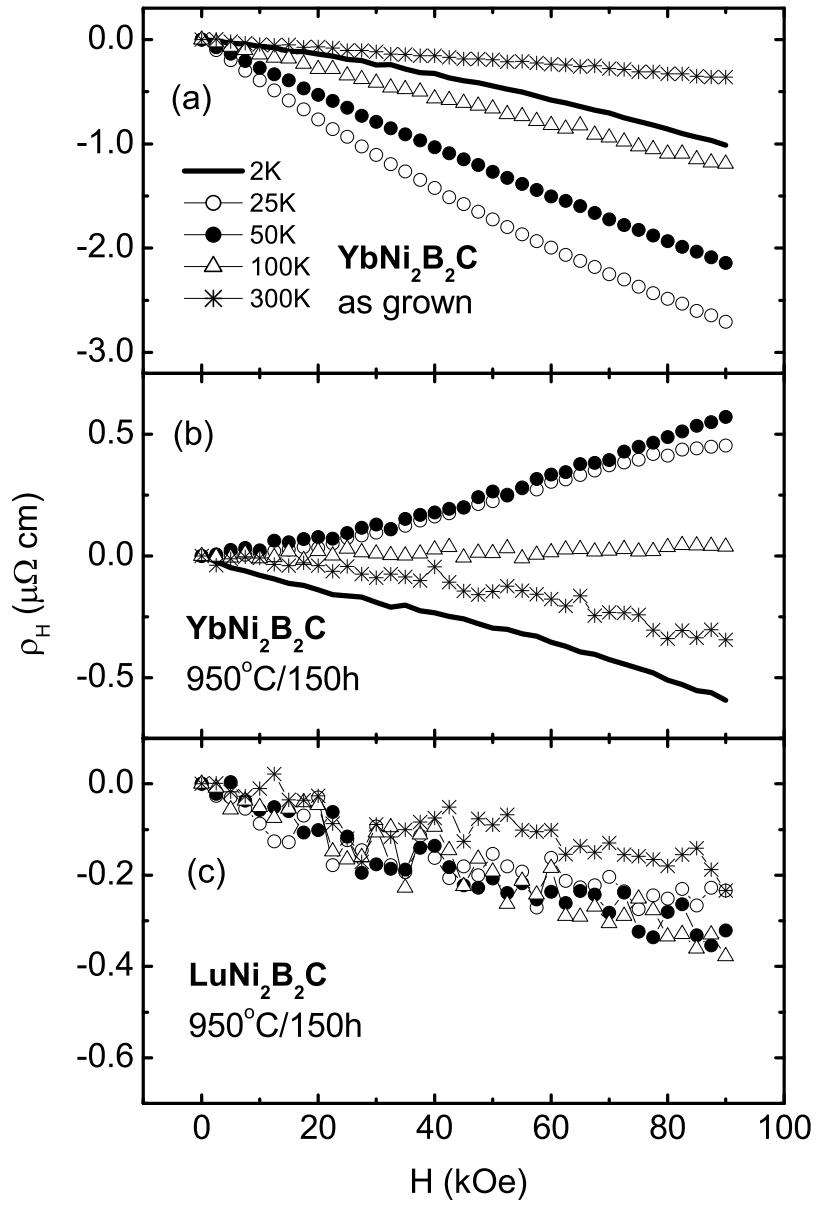


FIG. 2: Field-dependent Hall resistivity measured at different temperatures for (a) as grown $\text{YbNi}_2\text{B}_2\text{C}$; (b) annealed $\text{YbNi}_2\text{B}_2\text{C}$ and (c) annealed $\text{LuNi}_2\text{B}_2\text{C}$. Annealing conditions are shown on the respective panels, symbols are consistent for all four panels.

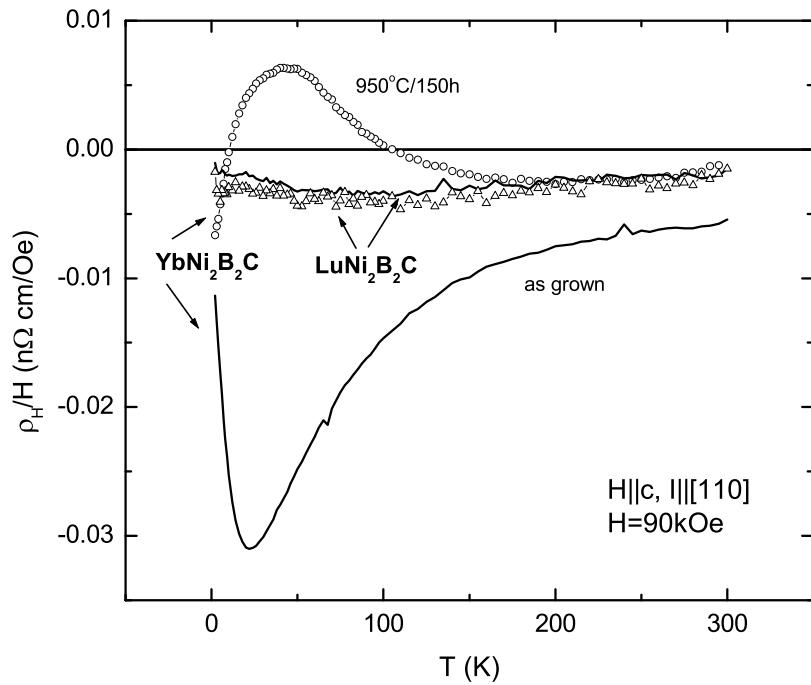


FIG. 3: The temperature-dependent Hall coefficient measured at $H = 90$ kOe for as-grown $\text{YbNi}_2\text{B}_2\text{C}$ (line) and $\text{LuNi}_2\text{B}_2\text{C}$ (line) and annealed at 950°C for 150 hours $\text{YbNi}_2\text{B}_2\text{C}$ (\circ) and $\text{LuNi}_2\text{B}_2\text{C}$ (\triangle) samples.

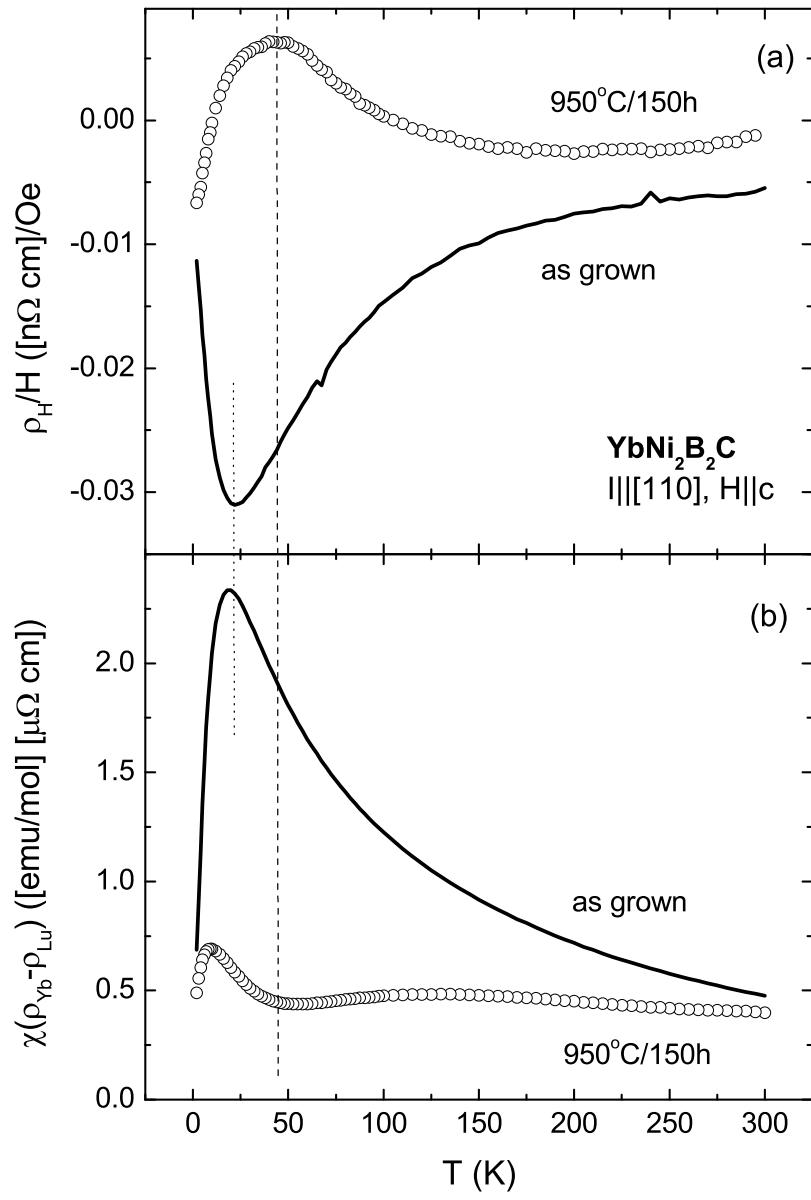


FIG. 4: Temperature-dependent (a) Hall coefficient measured at $H = 90$ kOe and (b) product of magnetic susceptibility and resonant scattering part of resistivity, $\chi(T)(\rho_{\text{Yb}}(T) - \rho_{\text{Lu}}(T))$ for as-grown (lines) and annealed at 950°C for 150 hours (○) $\text{YbNi}_2\text{B}_2\text{C}$ crystals. Dotted and dashed lines mark positions of the extrema in the curves for as-grown and annealed samples respectively.

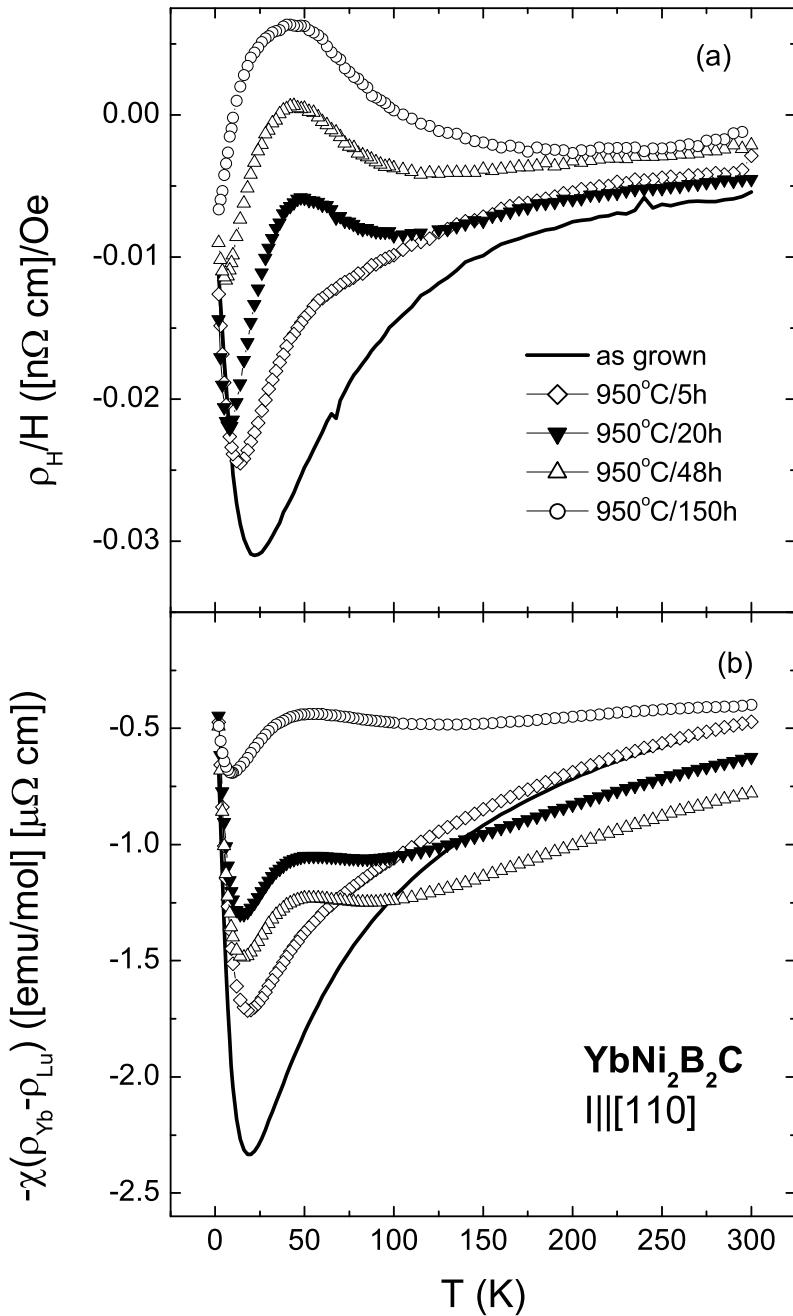


FIG. 5: Temperature-dependent (a) Hall coefficient measured at $H = 90$ kOe and (b) product of magnetic susceptibility and resonant scattering part of resistivity, $\chi(T)(\rho_{Yb}(T) - \rho_{Lu}(T))$ for as-grown (lines) and annealed at 950°C for 150 (○), 48 (△), 20 (▼) and 5 (◇) hours $\text{YbNi}_2\text{B}_2\text{C}$ crystals. Note: data in (b) are plotted as $-\chi(T)\rho_{magn}(T)$ for easier comparison with ρ_H/H data in (a).